

CLAIMS

1. A flame-retardant epoxy resin composition comprising an epoxy resin, a curing agent and a metal hydroxide,

wherein the curing agent is a phenolic resin (C) containing, in a molecular chain, a structural unit derived from a phenol (A) and a structural unit derived from an aromatic compound (B) other than the phenol (A).

2. A flame-retardant epoxy resin composition comprising an epoxy resin, a curing agent and a metal hydroxide,

wherein the epoxy resin is a novolac epoxy resin (D) obtained by glycidyletherifying a phenolic hydroxyl group of a phenolic resin (C) containing, in a molecular chain, a structural unit derived from a phenol (A) and a structural unit derived from an aromatic compound (B) other than the phenol (A).

3. A flame-retardant epoxy resin composition comprising an epoxy resin, a curing agent and a metal hydroxide,

wherein the curing agent is a phenolic resin

(C) containing, in a molecular chain, a structural unit derived from a phenol (A) and a structural unit derived from an aromatic compound (B) other than the phenol (A), and

the epoxy resin is a novolac epoxy resin (D) obtained by glycidyletherifying a phenolic hydroxyl group of a phenolic resin (C') containing, in a molecular chain, a structural unit derived from a phenol (A') and a structural unit derived from an aromatic compound (B') other than the phenol (A').

4. A flame-retardant epoxy resin composition according to Claim 1,

wherein the aromatic compound (B) is a compound selected from the group consisting of biphenyl and its derivatives, benzene and its derivatives, diphenyl ether and its derivatives, naphthalene and its derivatives, anthracene and its derivatives, fluorene and its derivatives, bisphenol fluorene and its derivatives, bisphenol S and its derivatives, bisphenol F and its derivatives and bisphenol A and its derivatives.

5. A flame-retardant epoxy resin composition according to Claim 2,

wherein the aromatic compound (B) is a compound selected from the group consisting of biphenyl and its derivatives, benzene and its derivatives, diphenyl ether and its derivatives, naphthalene and its derivatives, anthracene and its derivatives, fluorene and its derivatives, bisphenol fluorene and its derivatives, bisphenol S and its derivatives, bisphenol F and its derivatives and bisphenol A and its derivatives.

6. A flame-retardant epoxy resin composition according to Claim 3,

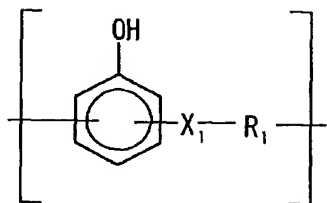
wherein the aromatic compound (B) is a compound selected from the group consisting of biphenyl and its derivatives, benzene and its derivatives, diphenyl ether and its derivatives, naphthalene and its derivatives, anthracene and its derivatives, fluorene and its derivatives, bisphenol fluorene and its derivatives, bisphenol S and its derivatives, bisphenol F and its derivatives and bisphenol A and its derivatives.

7. A flame-retardant epoxy resin composition according to Claim 1,

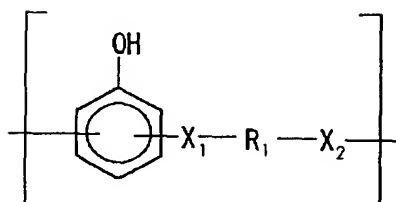
wherein the phenolic resin (C) has a recurring

unit represented by either of the following formulas

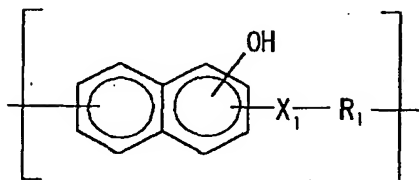
(I) to (IV):



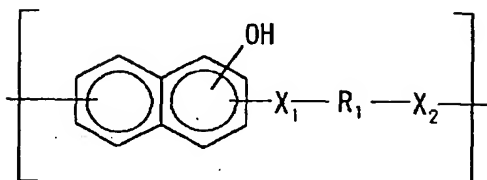
(I)



(II)



(III)



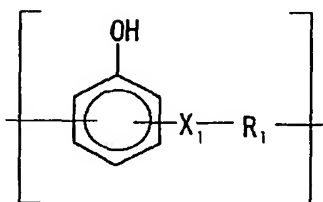
(IV)

(wherein X_1 and X_2 are each independently a C_{1-6} unsaturated chain structure linking group, or a C_{1-6} substituted or unsubstituted alkylene group; and R_1 is a phenylene group, a biphenylene group or a group derived from these groups).

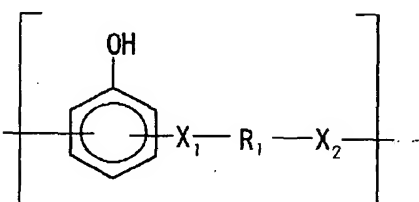
8. A flame-retardant epoxy resin composition according to Claim 2,

wherein the phenolic resin (C) has a recurring unit represented by either of the following formulas

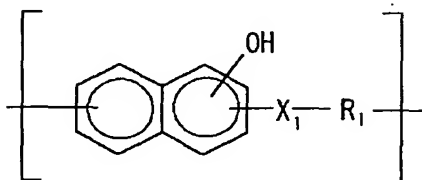
(I) to (IV):



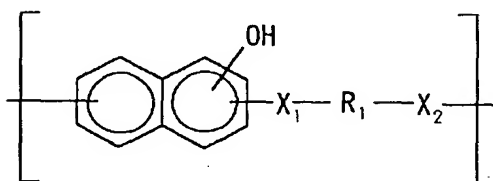
(I)



(II)



(III)

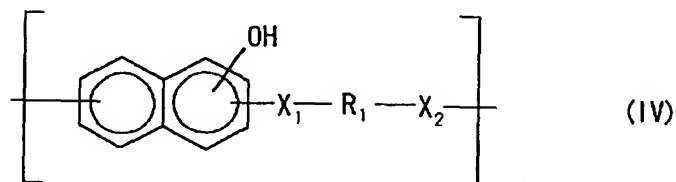
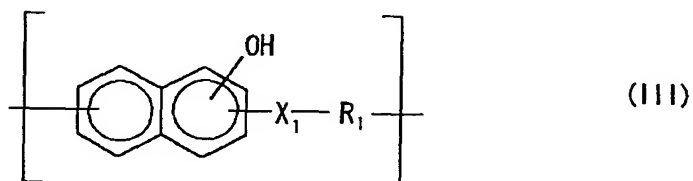
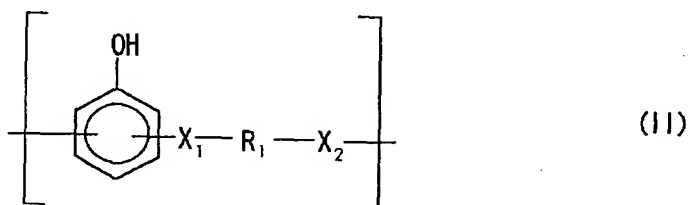
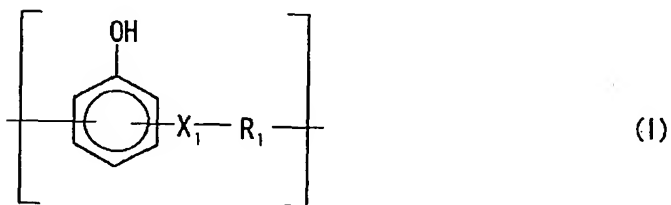


(IV)

(wherein X_1 and X_2 are each independently a C_{1-6} unsaturated chain structure linking group, or a C_{1-6} substituted or unsubstituted alkylene group; and R_1 is a phenylene group, a biphenylene group or a group derived from these groups).

9. A flame-retardant epoxy resin composition according to Claim 3,

wherein the phenolic resin (C) has a recurring unit represented by either of the following formulas (I) to (IV):



(wherein X_1 and X_2 are each independently a C_{1-6} unsaturated chain structure linking group, or a C_{1-6} substituted or unsubstituted alkylene group; and R_1 is a phenylene group, a biphenylene group or a group

derived from these groups).

10. A flame-retardant epoxy resin composition according to Claim 1,

wherein a content of the metal hydroxide is 10% by mass to 70% by mass relative to a total amount of the flame-retardant epoxy resin composition.

11. A flame-retardant epoxy resin composition according to Claim 2,

wherein a content of the metal hydroxide is 10% by mass to 70% by mass relative to a total amount of the flame-retardant epoxy resin composition.

12. A flame-retardant epoxy resin composition according to Claim 3,

wherein a content of the metal hydroxide is 10% by mass to 70% by mass relative to a total amount of the flame-retardant epoxy resin composition.

13. A flame-retardant epoxy resin composition according to Claim 1,

which further comprises a silicone compound of a branched structure main chain having an aromatic-derived group.

14. A flame-retardant epoxy resin composition according to Claim 2,

which further comprises a silicone compound of a branched structure main chain having an aromatic-derived group.

15. A flame-retardant epoxy resin composition according to Claim 3,

which further comprises a silicone compound of a branched structure main chain having an aromatic-derived group.

16. A flame-retardant epoxy resin composition according to Claim 13,

wherein a content of the metal hydroxide is 5% by mass to 70% by mass relative to a total amount of the flame-retardant epoxy resin composition.

17. A flame-retardant epoxy resin composition according to Claim 14,

wherein a content of the metal hydroxide is 10% by mass to 70% by mass relative to a total amount of the flame-retardant epoxy resin composition.

18. A flame-retardant epoxy resin composition according to Claim 15,

wherein a content of the metal hydroxide is 10% by mass to 70% by mass relative to a total amount of the flame-retardant epoxy resin composition.

19. A flame-retardant epoxy resin composition according to Claim 13,

wherein the silicone compound contains an unit (T unit) represented by the formula $\text{RSiO}_{1.5}$.

20. A flame-retardant epoxy resin composition according to Claim 14,

wherein the silicone compound contains an unit (T unit) represented by the formula $\text{RSiO}_{1.5}$.

21. A flame-retardant epoxy resin composition according to Claim 15,

wherein the silicone compound contains an unit (T unit) represented by the formula $\text{RSiO}_{1.5}$.

22. A flame-retardant epoxy resin composition according to Claim 13,

wherein the silicone compound contains a group reactive with the epoxy resin and/or the curing agent.

23. A flame-retardant epoxy resin composition according to Claim 14,

wherein the silicone compound contains a group reactive with the epoxy resin and/or the curing agent.

24. A flame-retardant epoxy resin composition according to Claim 15,

wherein the silicone compound contains a group reactive with the epoxy resin and/or the curing agent.

25. A flame-retardant epoxy resin composition according to Claim 22,

wherein the reactive group is hydroxyl group, C₁₋₅ alkoxy group, epoxy group or carboxyl group.

26. A flame-retardant epoxy resin composition according to Claim 23,

wherein the reactive group is hydroxyl group, C₁₋₅ alkoxy group, epoxy group or carboxyl group.

27. A flame-retardant epoxy resin composition according to Claim 24,

wherein the reactive group is hydroxyl group, C₁₋₅ alkoxy group, epoxy group or carboxyl group.

28. A flame-retardant epoxy resin composition according to Claim 1,

which is used for impregnation into a substrate and subsequent curing for formation of a laminate.

29. A flame-retardant epoxy resin composition according to Claim 2,

which is used for impregnation into a substrate and subsequent curing for formation of a laminate.

30. A flame-retardant epoxy resin composition according to Claim 3,

which is used for impregnation into a substrate and subsequent curing for formation of a laminate.

31. A flame-retardant epoxy resin composition according to Claim 1,

wherein the metal hydroxide is a metal oxide containing at least one element selected from the group consisting of aluminum, magnesium, zinc, boron, calcium, nickel, cobalt, tin, molybdenum, copper, iron and titanium.

32. A flame-retardant epoxy resin composition

according to Claim 2,

wherein the metal hydroxide is a metal oxide containing at least one element selected from the group consisting of aluminum, magnesium, zinc, boron, calcium, nickel, cobalt, tin, molybdenum, copper, iron and titanium.

33. A flame-retardant epoxy resin composition according to Claim 3,

wherein the metal hydroxide is a metal hydroxide containing at least one element selected from the group consisting of aluminum, magnesium, zinc, boron, calcium, nickel, cobalt, tin, molybdenum, copper, iron and titanium.

34. A flame-retardant epoxy resin composition according to Claim 31,

wherein the metal hydroxide is aluminum hydroxide, magnesium hydroxide or zinc borate.

35. A flame-retardant epoxy resin composition according to Claim 32,

wherein the metal hydroxide is aluminum hydroxide, magnesium hydroxide or zinc borate.

36. A flame-retardant epoxy resin composition according to Claim 33,

wherein the metal hydroxide is aluminum hydroxide, magnesium hydroxide or zinc borate.

37. An epoxy resin varnish solution obtained by dispersing the flame-retardant epoxy resin composition set forth in Claim 1, in an organic solvent.

38. An epoxy resin varnish solution obtained by dispersing the flame-retardant epoxy resin composition set forth in Claim 2, in an organic solvent.

39. An epoxy resin varnish solution obtained by dispersing the flame-retardant epoxy resin composition set forth in Claim 3, in an organic solvent.

40. A prepreg obtained by impregnating the flame-retardant epoxy resin composition set forth in Claim 1, into a substrate and curing the impregnated composition.

41. A prepreg obtained by impregnating the flame-retardant epoxy resin composition set forth in Claim 2, into a substrate and curing the impregnated composition.

42. A prepreg obtained by impregnating the flame-retardant epoxy resin composition set forth in Claim 3, into a substrate and curing the impregnated composition.

43. A laminate obtained by impregnating a flame-retardant epoxy resin composition comprising an epoxy resin, a curing agent and a metal hydroxide, into a substrate, curing the impregnated composition to obtain a prepreg, laminating a plurality of the prepregs, and hot-pressing them,

wherein the curing agent is a phenolic resin (C) containing, in a molecular chain, a structural unit derived from a phenol (A) and a structural unit derived from an aromatic compound (B) other than the phenol (A).

44. A laminate obtained by impregnating a flame-retardant epoxy resin composition comprising an epoxy resin, a curing agent and a metal hydroxide, into a

substrate, curing the impregnated composition to obtain a prepreg, laminating a plurality of the prepregs, and hot-pressing them,

wherein the epoxy resin is a novolac epoxy resin (D) obtained by glycidyletherifying a phenolic hydroxyl group of a phenolic resin (C) containing, in a molecular chain, a structural unit derived from a phenol (A) and a structural unit derived from an aromatic compound (B) other than the phenol (A).

45. A laminate obtained by impregnating a flame-retardant epoxy resin composition comprising an epoxy resin, a curing agent and a metal hydroxide, into a substrate, curing the impregnated composition to obtain a prepreg, laminating a plurality of the prepregs, and hot-pressing them,

wherein the curing agent is a phenolic resin (C) containing, in a molecular chain, a structural unit derived from a phenol (A) and a structural unit derived from an aromatic compound (B) other than the phenol (A), and

the epoxy resin is a novolac epoxy resin (D) obtained by glycidyletherifying a phenolic hydroxyl group of a phenolic resin (C') containing, in a molecular chain, a structural unit derived from a

phenol (A') and a structural unit derived from an aromatic compound (B') other than the phenol (A').

46. A laminate according to Claim 43,

wherein the aromatic compound (B) is a compound selected from the group consisting of biphenyl and its derivatives, benzene and its derivatives, diphenyl ether and its derivatives, naphthalene and its derivatives, anthracene and its derivatives, fluorene and its derivatives, bisphenol fluorene and its derivatives, bisphenol S and its derivatives, bisphenol F and its derivatives and bisphenol A and its derivatives.

47. A laminate according to Claim 44,

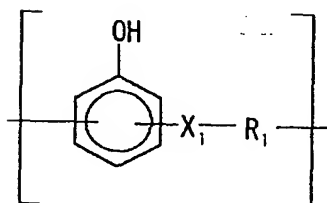
wherein the aromatic compound (B) is a compound selected from the group consisting of biphenyl and its derivatives, benzene and its derivatives, diphenyl ether and its derivatives, naphthalene and its derivatives, anthracene and its derivatives, fluorene and its derivatives, bisphenol fluorene and its derivatives, bisphenol S and its derivatives, bisphenol F and its derivatives and bisphenol A and its derivatives.

48. A laminate according to Claim 45,

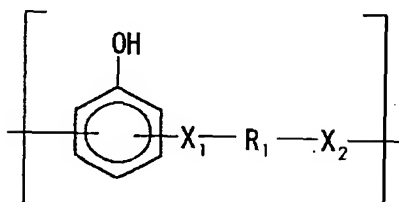
wherein the aromatic compound (B) is a compound selected from the group consisting of biphenyl and its derivatives, benzene and its derivatives, diphenyl ether and its derivatives, naphthalene and its derivatives, anthracene and its derivatives, fluorene and its derivatives, bisphenol fluorene and its derivatives, bisphenol S and its derivatives, bisphenol F and its derivatives and bisphenol A and its derivatives.

49. A laminate according to Claim 43,

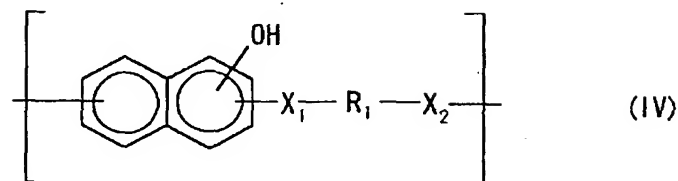
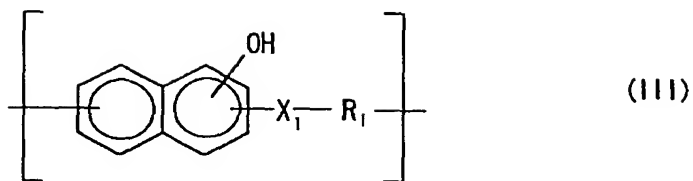
wherein the phenolic resin (C) has a recurring unit represented by either of the following formulas (I) to (IV):



(I)



(II)

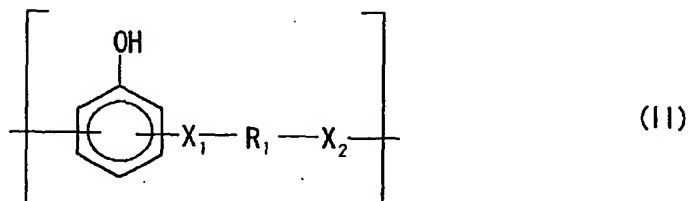
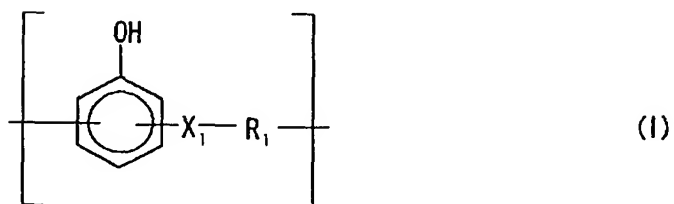


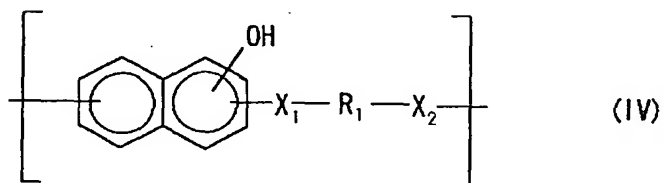
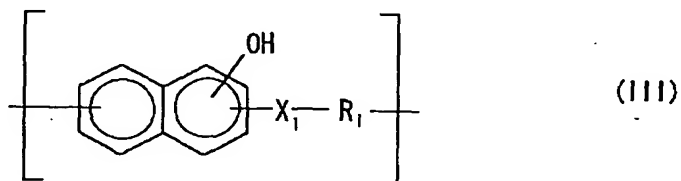
(wherein X_1 and X_2 are each independently a C_{1-6} unsaturated chain structure linking group, or a C_{1-6} substituted or unsubstituted alkylene group; and R_1 is a phenylene group, a biphenylene group or a group derived from these groups).

50. A laminate according to Claim 44,

wherein the phenolic resin (C) has a recurring unit represented by either of the following formulas

(I) to (IV):

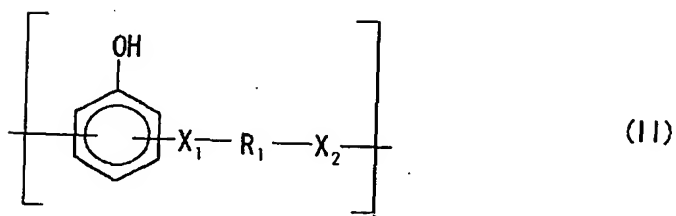
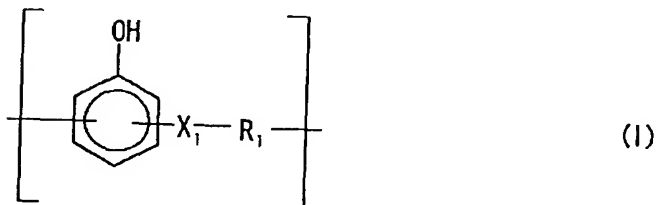


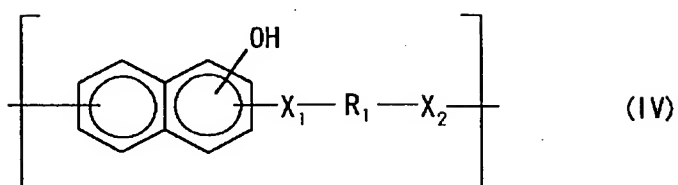
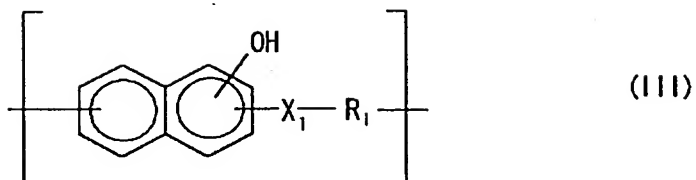


(wherein X_1 and X_2 are each independently a C_{1-6} unsaturated chain structure linking group, or a C_{1-6} substituted or unsubstituted alkylene group; and R_1 is a phenylene group, a biphenylene group or a group derived from these groups).

51. A laminate according to Claim 45,

wherein the phenolic resin (C) has a recurring unit represented by either of the following formulas (I) to (IV):





(wherein X_1 and X_2 are each independently a C_{1-6} unsaturated chain structure linking group, or a C_{1-6} substituted or unsubstituted alkylene group; and R_1 is a phenylene group, a biphenylene group or a group derived from these groups).

52. A laminate according to Claim 43,

wherein a content of the metal hydroxide is 10% by mass to 70% by mass relative to a total amount of the flame-retardant epoxy resin composition.

53. A laminate according to Claim 44,

wherein a content of the metal hydroxide is 10% by mass to 70% by mass relative to a total amount of the flame-retardant epoxy resin composition.

54. A laminate according to Claim 45,
wherein a content of the metal hydroxide is 10%
by mass to 70% by mass relative to a total amount of
the flame-retardant epoxy resin composition.

55. A laminate according to Claim 43,
which further comprises a silicone compound of
a branched structure main chain having an aromatic-
derived group.

56. A laminate according to Claim 44,
which further comprises a silicone compound of
a branched structure main chain having an aromatic-
derived group.

57. A laminate according to Claim 45,
which further comprises a silicone compound of
a branched structure main chain having an aromatic-
derived group.

58. A laminate according to Claim 43,
wherein a content of the metal hydroxide is 5%
by mass to 70% by mass relative to a total amount of
the flame-retardant epoxy resin composition.

59. A laminate according to Claim 44,

wherein a content of the metal hydroxide is 5% by mass to 70% by mass relative to a total amount of the flame-retardant epoxy resin composition.

60. A laminate according to Claim 45,

wherein a content of the metal hydroxide is 5% by mass to 70% by mass relative to a total amount of the flame-retardant epoxy resin composition.

61. A laminate according to Claim 43,

wherein the silicone compound contains an unit (T unit) represented by the formula $\text{RSiO}_{1.5}$.

62. A laminate according to Claim 44,

wherein the silicone compound contains an unit (T unit) represented by the formula $\text{RSiO}_{1.5}$.

63. A laminate according to Claim 45,

wherein the silicone compound contains an unit (T unit) represented by the formula $\text{RSiO}_{1.5}$.

64. A laminate according to Claim 43,

wherein the silicone compound contains a group

reactive with the epoxy resin and/or the curing agent.

65. A laminate according to Claim 44,
wherein the silicone compound contains a group
reactive with the epoxy resin and/or the curing agent.

66. A laminate according to Claim 45,
wherein the silicone compound contains a group
reactive with the epoxy resin and/or the curing agent.

67. A laminate according to Claim 43,
wherein the reactive group is hydroxyl group,
C₁₋₅ alkoxy group, epoxy group or carboxyl group.

68. A laminate according to Claim 44,
wherein the reactive group is hydroxyl group,
C₁₋₅ alkoxy group, epoxy group or carboxyl group.

69. A laminate according to Claim 45,
wherein the reactive group is hydroxyl group,
C₁₋₅ alkoxy group, epoxy group or carboxyl group.

70. A laminate according to Claim 43,
wherein the metal hydroxide is a metal oxide
containing at least one element selected from the

group consisting of aluminum, magnesium, zinc, boron, calcium, nickel, cobalt, tin, molybdenum, copper, iron and titanium.

71. A laminate according to Claim 44,

wherein the metal hydroxide is a metal oxide containing at least one element selected from the group consisting of aluminum, magnesium, zinc, boron, calcium, nickel, cobalt, tin, molybdenum, copper, iron and titanium.

72. A laminate according to Claim 45,

wherein the metal hydroxide is a metal oxide containing at least one element selected from the group consisting of aluminum, magnesium, zinc, boron, calcium, nickel, cobalt, tin, molybdenum, copper, iron and titanium.

73. A laminate according to Claim 43,

wherein the metal hydroxide is aluminum hydroxide, magnesium hydroxide or zinc borate.

74. A laminate according to Claim 44,

wherein the metal hydroxide is aluminum hydroxide, magnesium hydroxide or zinc borate.

75. A laminate according to Claim 45,
wherein the metal hydroxide is aluminum
hydroxide, magnesium hydroxide or zinc borate.

76. A laminate according to Claim 43,
which satisfies the following conditions (a) to
(d):

(a) $45 \leq \sigma \leq 100$, $3 \leq E \leq 12$

[wherein σ is a bending strength (MPa) of the
laminate at $230 \pm 10^\circ\text{C}$ and E is a flexural modulus
(GPa) of the laminate at $230 \pm 10^\circ\text{C}$],

(b) $30 \leq G \leq 60$

[wherein G is a proportion (mass %) of the substrate
in a total amount of the laminate],

(c) $F \leq 45$ (mass %), F (mass %) = $R \times 100/X$

(wherein R is an amount of a thermal decomposition
product other than water, generating from a room
temperature to 500°C , and X is a content of the resin
in the laminate), and

(d) $4 \leq V \leq 13$

[wherein V is an amount (V mass %) of a water vapor
generating from a room temperature to 500°C , relative
to a total amount of the laminate, when the laminate
is subjected to thermal decomposition at a

temperature elevation rate of 10°C/min at an air flow rate of 0.2 liter/min].

77. A laminate according to Claim 44,

which satisfies the following conditions (a) to

(d):

(a) $45 \leq \sigma \leq 100$, $3 \leq E \leq 12$

[wherein σ is a bending strength (MPa) of the laminate at $230 \pm 10^\circ\text{C}$ and E is a flexural modulus (GPa) of the laminate at $230 \pm 10^\circ\text{C}$],

(b) $30 \leq G \leq 60$

[wherein G is a proportion (mass %) of the substrate in a total amount of the laminate],

(c) $F \leq 45$ (mass %), F (mass %) = $R \times 100/X$

(wherein R is an amount of a thermal decomposition product other than water, generating from a room temperature to 500°C , and X is a content of the resin in the laminate), and

(d) $4 \leq V \leq 13$

[wherein V is an amount (V mass %) of a water vapor generating from a room temperature to 500°C , relative to a total amount of the laminate, when the laminate is subjected to thermal decomposition at a temperature elevation rate of $10^\circ\text{C}/\text{min}$ at an air flow rate of 0.2 liter/min].

78. A laminate according to Claim 45,

which satisfies the following conditions (a) to

(d):

(a) $45 \leq \sigma \leq 100$, $3 \leq E \leq 12$

[wherein σ is a bending strength (MPa) of the laminate at $230 \pm 10^\circ\text{C}$ and E is a flexural modulus (GPa) of the laminate at $230 \pm 10^\circ\text{C}$],

(b) $30 \leq G \leq 60$

[wherein G is a proportion (mass %) of the substrate in a total amount of the laminate],

(c) $F \leq 45$ (mass %), F (mass %) = $R \times 100/X$

(wherein R is an amount of a thermal decomposition product other than water, generating from a room temperature to 500°C , and X is a content of the resin in the laminate), and

(d) $4 \leq V \leq 13$

[wherein V is an amount (V mass %) of a water vapor generating from a room temperature to 500°C , relative to a total amount of the laminate, when the laminate is subjected to thermal decomposition at a temperature elevation rate of $10^\circ\text{C}/\text{min}$ at an air flow rate of 0.2 liter/min].